Removal of thermophilic spores from gum Arabic streams using ceramic alumina microfiltration membranes

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ABSTRACT

The microfiltration (MF) of high solids content gum arabic solutions (15 wt%) inoculated with Bacillus mycoides *spores (10^5 CFU ml^{-1}) was carried out using* Membralox *tubular ceramic membranes with a nominal pore size of 0.8 µm. Consistent permeate fluxes were achievable over multiple fouling and cleaning cycles, while giving low rejection of solids and high rejection of spores (after ten cycles, a permeate flux of 42.9 Lm^{-2} h^{-1}, a solids retention of 19.8%, and a spore rejection of 5.0 log orders were achieved). Although fouling during filtration was severe, permeate fluxes could be restored to a satisfactory condition after cleaning with 0.5 wt% NaOH solution containing 200 ppm NaOCl. Results were described by a two species first order removal model, whereby one species was removed quickly by cleaning and the other was more difficult to remove. The optimum cleaning time for NaOH + NaOCl solutions at 60° C was found to be* ca. *20 minutes. Subsequent citric acid cleans had a negative effect upon restoring permeate flux.*

INTRODUCTION

Gum Arabic (GA) is the dried exudation obtained from various species of *Acacia* trees of the leguminosae family. About 500 species of *Acacia* are distributed across tropical and sub-tropical areas of Africa, India, Australia, Central America and southwest North America, but only a few are commercially important. Most production occurs in the 'gum belt' proper of central and western Sudan where the trees *Acacia Senegal* grow in sandy soils under water scarce conditions. GA is the country's second largest export and Sudan provides 85 % of the world supply of GA, which is predominantly used in the food and beverage industry for its emulsifying and stabilising properties.

Gum arabic is a slightly acidic complex polysaccharide that consists of three main components; (i) arabinogalactan (AG) with a molar mass of approximately 280 kg mol^{-1} that

makes up ≈88 wt%, (ii) arabinogalactan-protein (AGP) complex which accounts for ≈10 wt.%, and has a molar mass of 1500 kg mol^{-1}, (iii) glycoprotein (GI) which makes up the final ≈2 wt.% and has a molar mass of 250 $kgmol^{-1}$. The AGP complex is the active component responsible for GA's emulsifying and stabilising properties (Fauconnier et al. 2000, Manning & Bird 2015, Manning et al 2016).

It is well known that conditions in which GA is grown and collected are susceptible to contamination by bacterial spores such as *Bacillus cereus* (Farag Zaied et al. 2007); a known food-poisoning bacterium which is resilient to thermal pasteurisation. Removal of such spores is thus difficult by heat treatment alone given the thermo-resistant properties and resulting degradation of product quality with excessive temperatures (Decloux et al. 1996). Therefore the use of a filtration method to act as a cold pasteurisation technique is an attractive alternative.

In this study, microfiltration was used as a method of cold pasteurisation. Reconstituted spray-dried gum solutions were inoculated with a microbial load of *Bacillus mycoides* spores (maximum diameter 1.53+/- 0.18 μm (Head & Bird 2013 b) as an analogue to *B. cereus*. The gum solutions were then micro-filtered over multiple fouling and cleaning cycles with the objective of achieving high permeate flux, high gum solids transmission and high spore retention over a period of time, and measuring the effect of membrane ageing on filtration performance. The overall aim of the experiments was to develop an industrially relevant protocol to modify industrial practice. Feeds, processing conditions and cleaning agents were all selected according to previous industrial practice.

The removal of bacteria from food and dairy products using microfiltration is a well-established and proven technology (Pafylias et al. 1996; Tomasula et al. 2011). However, typical dairy microfiltration processes treat low solids content aqueous streams; skimmed milk has a protein content of approximately 3.5wt% (Piry et al. 2008). Higher solids content streams such as those used in the processing of milk protein isolate are more difficult to filter effectively due to higher viscosities and higher rates of membrane fouling leading to low flux and solids transmission (Head & Bird 2013 a; Head & Bird 2013 b). Similar difficulties were expected with high solids content GA feeds.

MATERIALS

Experimental apparatus

The experiments were carried out on a laboratory scale cross-flow microfiltration apparatus as described by Head & Bird (2013b). The apparatus consisted of two circulation loops. A centrifugal pump circulated feed from the tank (20 L) through a heater to raise the feed to the desired temperature, and then by adjusting control valves the feed was diverted to the retentate circulation loop. The retentate was partially recycled through a cooler to the retentate circulation pump. Trans-membrane pressure, temperature and cross-flow velocity were measured by an arrangement of pressure transducers, a thermocouple (Cole-Parmer) and electromagnetic flow meter (MAG Magflo 1100). Permeate flux measurements were logged automatically to a computer.

Membranes

The membrane tested was a Pall Membralox™ (Pall, USA) tubular ceramic membrane. The membrane had a nominal pore size of 0.8 μm, 19 cylindrical channels, 1020 mm long and 4 mm in diameter giving an active membrane area on each membrane of 0.24 m^2. However, to achieve high cross flow velocities on the same experimental apparatus, only 3 channels were used by blocking off 16 channels using stainless steel backed plugs and gaskets, giving an active membrane area of 0.04 m^2. By pure water flux measurements, this was shown to a suitable method of increasing cross-flow velocity for laboratory scale measurements without increasing the size of the related apparatus.

Gum arabic fouling suspension

The gum arabic used was supplied by *Kerry group plc.*as a spray-dried powder. For each experiment, 16 kg of solution was made up the previous day by mixing the gum powder with reverse osmosis treated water (measured conductivity of 8.0 μS cm^{-1}) at room temperature and stirred continuously for 2 hours. The solution was left overnight at room temperature to ensure complete gum dissolution. When diluted with holding water in the rig a feed of 15wt% GA solids resulted.

Bacillus mycoides spore inoculation

The method used to prepare a spore solution of *Bacillus mycoides* is described by Seale et al (2008). Ten millilitres of the spore solution was pipetted to the appropriate feeds before experiments to give a retentate microbial load of *ca*. 10^5 CFU ml^{-1}. The spores are ellipsoid in shape with a width of 0.9 ± 0.11 μm and length of 1.53 ± 0.18 μm (Head & Bird 2013b).

Cleaning reagents

Three cleaning reagents were used. The first chemical cleaning stage used a solution of 0.5% w/w sodium hydroxide (NaOH) with 200 ppm sodium hypochlorite (NaOCl) added. The NaOH concentration chosen was that determined by Bird & Bartlett (1995) to be the most effective for proteinaceous deposit removal. The presence of NaOCl was included for two reasons, (i) It is a widely used and effective sanitiser even at low concentrations (Young and Setlow 2003) and (ii) NaOH solutions alone proved to be ineffective at restoring acceptable permeate flux after gum arabic fouling. It was found that adding 200ppm NaOCl was effective in increasing flux recovery. In the microfiltration of rough beer Gan et al (1999) demonstrated that combining caustic and oxidation cleans was more effective than running them in two separate stages. Gan et al (1999) also found a flux recovery associated with an acid clean following alkali cleaning of a ceramic membranes. Likewise the second cleaning stage in these experiments used 0.1% w/w citric acid solution.

METHODS

Each cycle consisted of seven stages; Initial pure water flux, fouling, a first rinse, NaOH + NaOCl cleaning, second rinse, citric acid cleaning and then a final rinse. Each cycle completed a full fouling and cleaning of the membrane, with the first of 10 successive cycles

was carried out on a pristine membrane. The full conditions of each stage are given in Table 1. A typical profile of total resistance over one fouling and cleaning cycle can be observed in Figure 1. Resistance increases during fouling for the first 60 minutes, with subsequent rinsing and cleaning stages removing fouling and lowering resistances (although the apparent resistance during resistance is higher than that seen during fouling - as discussed in the *Results and Discussion* section.

Table 1. Operating conditions for stages in complete fouling and cleaning cycle

Stage	Duration min	CFV ms^{-1}	TMP bar	Temperature ° C	Measured Resistance m^{-1}
Initial Pure water flux	5	8.5	2.5	20	R_m
Fouling	60	8.5	2.9	50	R_t
Rinsing	30	8.5	0.5	20	R_f
NaOH + NaOCl cleaning	30	11	2.5	60	$R_{fc'}$
Rinsing	5	8.5	2.5	20	$R_{n'}$
Citric acid cleaning	30	11	2.5	60	R_{fc}
Rinsing	5	8.5	2.5	20	R_n

Initial pure water flux (PWF)

Resistance to permeate flux could be measured at each stage of the cycle. Initial resistance by the pristine membrane (R_{m1}) was measured in the first stage of cycle one by measuring the initial pure water flux and using Equation 1:

$$J = \frac{TMP}{\mu R_m} \qquad \text{Equation 1}$$

In the same way, the resistance of the cleaned membrane (R_n) was calculated by taking the flux J_n and substituting R_m with R_n.

Fouling

Fouling of the membrane was carried out by first heating the gum arabic feed to 50°C, then recirculating it through the membrane apparatus to give a cross flow velocity (CFV) through the membrane of 8.5 ms^{-1}. The feed was kept at 50°C and the membrane fouled for 60 minutes at a TMP of 2.5 bar. This was the highest possible CFV with the minimum TMP at that CFV, a simple choice of conditions shown to be optimum in the microfiltration of gum arabic by Decloux et al (1996) and widely reported in the literature for other microfiltration

processes (Gésan et al 1993).. The resistance in series model was applied to calculate the fouling resistance, namely:

$$J = \frac{TMP}{\mu(R_m + R_f)} \qquad \text{Equation 2}$$

Where R_f is the fouling resistance at cycle n.

Several authors have previously used relative flux decline (RFD) as a means to describe membrane performance during fouling (Weis et al. 2003; Blanpain-Avet et al. 2004). RFD is a ratio between permeate flux at time 0 (J_0) and final fouling flux at the end of the fouling step (J_{60}). The equation used to calculate RFD is:

$$RFD = \left(1 - \frac{J_f}{J_0}\right) \qquad \text{Equation 3}$$

Cleaning

After each fouling cycle, a pure water rinse, a NaOH + NaOCl clean and a citric acid clean were carried out. A CFV of 8.5m s^{-1} and a TMP of 2.5 barwere applied as operating conditions, while still maintaining a positive TMP along the entire membrane. Although Bartlett et al (1995) found that cleaning with zero TMP was more efficient than with a positive TMP, no flux data can be measured during cleaning, hence a positive TMP was selected.

Hydraulic cleanliness of the membrane was measured by the hydraulic cleanliness coefficient (HCC), as defined by the following:

$$HCC = \frac{(R_n - R_m)}{R_m} \qquad \text{Equation 4}$$

here R_m is the clean membrane resistance and R_n is the resistance to pure water after cleaning. According to Blanpain-Avet et al (2004) HCC < 0.05 is considered hydraulically clean as is within the limits of acceptable error in flux measurements.

Solids retention

Refractive index was determined by using a Reichert r^2 Mini handheld refractometer. A good correlation was found between the refractive index of dissolved product solids content. The correlation as shown in Equation 5 was determinedby measuring the refractive index of

different solids content samples and comparing them to dry mass (DM) measurements of samples weighed and then dried in an oven at 55 °C until a constant mass was recorded.

$$DM\ (\%\, w/w) = 0.00967.Brix^{\circ} \qquad \text{Equation 5}$$

Solids Retention was determined as follows:

$$SR = \left(1 - \frac{DM_P}{DM_F}\right) \qquad \text{Equation 6}$$

here DM_P and DM_F are the dry masses of the permeate and the feed samples respectively $Brix^{\circ}$ measures the sugar content of the aqueous solution.

Clarity

Clarity was measured by a turbidity measurement ratio of feed and permeate samples after 60 minutes of filtration. Feed and permeate were both diluted to 10 % w/w solids and turbidity taken using a HANNA HI 93703 Portable Microprocessor Turbidity Meter at room temperature. Clarity was calculated as follows:.

$$C_s = \left(1 - \frac{\tau_p}{\tau_f}\right) \qquad \text{Equation 7}$$

Here τ_f and τ_p are the turbidity values in NTU of feed and permeate samples respectively.

Spore enumeration

Retentate and permeate total spore counts were performed for each filtration. The method used was the Petrifilm™ aerobic film plating technique as previously described by Bowen et al (2002). The total spore counts of each sample are presented as Log_{10} CFU ml^{-1}.

RESULTS AND DISCUSSION

Membrane performance during fouling

Membrane performance over the 10 fouling cycles was measured by quasi-steady state permeate flux at the end of fouling (J_t), the corresponding quasi-steady state total resistance at the end of fouling (R_t), the total irreversible fouling (R_f) and the relative flux decline (RFD). Feed and permeate samples were taken at the end of fouling to determine solids retention (SR), clarity (C_s) and permeate spore count.

A variation of J_t was observed to be between 15 and 56 $Lm^{-2}h^{-1}$, with an average of 35.3 $Lm^{-2}h^{-1}$. R_t and R_f were observed over the course of the 10 cycles and little trend in membrane performance was apparent (Figure 2). This is consistent with Blanpain-Avet et al (2004) who

carried out multiple fouling and cleaning cycles on a ceramic membrane using whey protein isolate as a model fouling suspension, finding little variation in R_f and R_t over 10 cycles.

Figure 3 shows progressively decreasing relative flux decline (RFD), with RFD decreasing from a maximum of 98.9% in the first cycle to a minimum of 94.4% in the tenth cycle. This is different from Blanpain-Avet et al (2004), who found no trend in RFD, and is contrary to the work reported by Bird et al (2003), who found that multiple fouling of polymeric UF membranes with spent sulphite liquor showed a gradual increase in RFD over 15 cycles. In the current work, the decreasing RFD value combined with no apparent trend in R_t, suggests that ineffective cleaning protocols were used, leaving deposits on the membrane surface. It is hypothesised that because R_n does not affect R_t, the deposits left on the membrane are affecting surface-particle interactions rather than blocking pores. Arabinogalactan polysaccharides that are responsible for over 90% of the gum molecular weight are hydrophilic. Evans and Bird (2008) showed that for a hydrophilic polysulphone membrane, multiple cleans after fouling of black tea liquor decreased pure water flux while hydrophobicity was decreased. It is probable that similar phenomena are being observed with gum arabic and the Al_2O_3 membranes, although the tubular channels prevented contact angle analysis to confirm this hypothesis. In every fouling cycle, it was observed that the total resistance during rinsing (R_t) measured by pure water flux was significantly higher than the total resistance observed by measuring the permeate flux of the gum solution. The practical effect of this is GA solutions showing favourable mass transfer compared to that of water. Rabiller-Baudry et al (2005) found that electrostatic interactions played a major role in fouling and limited flux during the ultrafiltration of skimmed milk. Zhang et al (2007) showed that attachment of GA onto γ-Al_2O_3 nano-particles reduced their zeta-potential, and prevented agglomeration by repulsive forces. This can explain why it is possible for a thin layer of fouling on the membrane to actually limit further fouling by repelling the high solids content of the GA feed.

Solids retention (SR) did not appear to show significant correlation between cycle number and value, however there was a moderately positive correlation between SR and R_t in Figure 4. This relationship is significant when calculating the eventual dry mass of GA produced as increased solids retention exacerbates a low permeate flux. For example cycle 2 (J_t = 56.5 $Lm^{-2}h^{-1}$ and SR = 9.3 %) resulted in a final flux of GA solids of 8.1 kg m^{-2} h^{-1}, whereas cycle 5 (J_t = 15.3 $Lm^{-2}h^{-1}$ and SR = 40.1 %) resulted in a GA solids flux of 1.5 kg m^{-2} h^{-1}. This example shows steady state flux in cycle 2 being 3.7 times that of cycle five. However, because of the increased selectivity at lower permeate flux cycle 5 produced 5.6 times the amount of GA product compared to that of cycle 2.

Permeate clarity (C_s) was high throughout the trial and varied little between 78.9 % and 88.3 % over the 10 cycles (Table 2). There was little correlation between C_s and cycle number or R_t. The high clarity is consistent with that found by Decloux et al. (1996) in their experiments for the microfiltration of GA.

Membrane performance proved to be very positive with respect to feed sterilisation. When the feed was first inoculated with ca.10^5 CFU ml^{-1} of *B. mycoides* spores, permeate spore

counts found no trace of mycoides spores in all but two cycles. Similar performance of the same membranes using the same spores was recorded by Head and Bird (2013b) while using microfiltration to treat high solids content milk protein isolate feeds.

Table 2. Summary of GA permeate flux, solids retention (SR), clarification and permeate spore count

Cycle	J_t $Lm^{-2}h^{-1}$	SR %	C_s %	Permeate spore count log CFU ml^{-1}
1	35.1	3.7	78.9	0.00
2	56.5	9.3	82.1	2.22
3	27.8	17.3	86.9	0.00
4	32.3	17.9	85.7	0.00
5	15.3	40.1	88.3	0.00
6	25.6	19.1	87.1	0.00
7	23.9	21.0	85.5	0.00
8	47.5	14.8	86.7	1.82
9	45.8	14.8	86.0	0.00
10	42.9	19.8	87.2	0.00

Membrane cleaning efficiency

Figure 5 presents flux recovery (FR) over the 10 cycles. FR is defined as PWF for cycle n as a fraction of PWF for cycle n+1. It was observed that the FR does not decrease significantly after 10 cycles, meaning cleaning efficiency is not affected by the ageing of the membrane. The trend observed is a gradual increase in FR between cycle 1 and 6, generally below 100 % FR but peaking at 194 %. FR then gradually decreased to 43 % in cycle 10 compared to that of 54 % in cycle one. A 'one-off' cycle observation of higher than 100 % FR has been observed previously by Blanpain-Avet et al. (2004) in the microfiltration of whey protein concentrate through ceramic membranes. An inefficient cleaning regime may have failed to completely solubilise foulant that has built up on the virgin membrane during cycle one. The modified surface would then discourage fouling in subsequent cycles, leading to an increased FR, that is, the cleaning appears more efficient as less new foulant has built up than in previous cycles. It has been suggest by Blanpain-Avet et al (2004) that the significant jump to an FR of far over 100 % (as also observed in Figure 5) can be caused by an uneven removal of swollen AGP agglomerates from the membrane surface present from the previous cleaning cycles. A hydraulic cleanliness criterion (HCC) is shown in Figure 6 following both NaOH + NaOCl cleaning and after a subsequent citric acid clean. The range of HCC varies between 1.52 and -0.61 with no apparent trend over the 10 cycles. For six of the cycles HCC > 0, that is, hydraulic cleanliness was not achieved. It can clearly be seen that citric acid has a detrimental effect on membrane cleanliness as HCC is greater after every citric acid clean

compared to that after the initial NaOH + NaOCl clean, an effect further observed in Figure 7.

Effect of rinsing, NaOH + NaOCl and citric acid on membrane cleaning

As observed in a typical profile of total resistance over one fouling and cleaning cycle (Figure 1) resistance increases over fouling for the first 60 minutes, then rinsing removes a certain amount of reversible fouling.NaOH + NaOCl cleaning then removes irreversible fouling, followed by the eventual citric acid clean, which appears to increase membrane resistance. To quantify this increase in resistance during citric acid cleaning, Figure 7 shows $R_n/R_{n'}$ over each cycle. This ratio ranges from 0.97 to 3.10 and is consistently above 1 in all but cycle 9, leading to the conclusion that all effective cleaning is carried out at the NaOH + NaOCl cleaning stage and therefore the citric acid cleaning step is effectively redundant and possibly harming the recovery of membrane performance.*Kinetics of NaOH + NaOCl cleaning*

Cleaning models are useful in determining the necessary membrane cleaning duration and the efficiency of other cleaning parameters such as temperature and chemical concentration. They can also provide evidence of the nature of membrane fouling. NaOH + NaOCl cleaning is the active cleaning stage responsible for the restoration of the otherwise irreversibly fouled membrane; therefore this stage was considered when investigating cleaning kinetics.

Cleaning is characterised by an initial fast removal of foulant, decreasing resistance at a high rate, followed by slower removal of foulant, seen as a more gradual decrease towards the end of the cleaning cycle (Figure 8). This trend in cleaning has been reported previously for the cleaning of ceramic membranes by other authors (Bartlett et al. 1995; Blanpain-Avet et al. 2009). Resistance changes provide an indication of surface condition, but do not relate directly to either the removal or reaction of an individual fouling species. Nevertheless, resistance is an extremely useful practical measure of a membrane s performance.

Zero and first order reactions have been modelled to experimental resistance data over time for the NaOH + NaOCl cleaning cycle (SigmaPlot, Systat Software, San Jose, CA). The reaction of a single species fits an *nth* reaction order according to Equation 8. A first order reaction (n=1) implies there is instant penetration of the chemical agent through the fouling surface and a zero order reaction (n=0) implies the removal rate of fouling is constant at any time and is independent of fouling thickness.

$$\frac{dR_{fc}}{dt} = -k.R_{fc}^{n} \qquad \text{Equation 8}$$

Where R_{fc} is the initial fouling resistance at the end of rinsing that is subsequently removed by cleaning and k is the removal rate constant.,

An example of the curve fit is given in Figure 8, and the coefficient of determination (r^2) for each curve over cycles 3 – 10 were calculated. It is immediately visible that a one species zero order reaction is not occurring, as the cleaning is not linear with time, this is characterised by a mean r^2 of 0.336 over the seven cycles measured. However, a one species first order model provides a satisfactory fit, with a mean r^2 value of 0.958 and shows a mean removal rate constant (k) of 1.83 ±.83 min^{-1}.

There are two possible mechanistic explanations for first order hydraulic resistance decline recorded. One is that the deposit morphology is an open one – facilitating an intimate contact between the cleaning agent and the entire depth of the fouling deposit. An alternative explanation is that the deposit is not confluent, and a uniform film, but effectively consists of islands of deposition. Thus a zero order removal of an uneven deposition mass could give the appearance of first order removal overall, and a first order decline in the hydraulic resistance.

CONCLUSIONS

The multiple fouling and cleaning of a 0.8 μm alumina ceramic membrane has been undertaken for feeds comprising of 15% w/w solutions of gum arabic containing *Bacillus mycoides* spores. Cleaning was carried out using 0.5 % w/w solutions of NaOH containing 200ppm of sodium hypochlorite, followed by a 0.1 % w/w citric acid clean. Fouling was carried out at 50° C and cleaning at 60° C. Fouling filtration performance was encouraging, with reasonable permeate flux being maintained after 10 cycles (cycle 10 J_{60} = 42.9 $Lm^{-2}h^{-1}$). The membrane consistently performed well at spore rejection, with log reduction values up to 5.0 log_{10} CFU ml^{-1}; while solids retention varied between 3.7 and 40.1%, but was correlated with observed permeate flux, indicating that maintaining high permeate flux is critical. The gradual decline in membrane recovery that has been previously observed for fouling of polymeric membranes (Weis and Bird 2001) was not observed here. However a relatively consistent performance was seen after the first three cleaning cycles, in common with previous work on multiple cycle ceramic membrane fouling (Blanpain-Avet et al. 2004).

Complete removal of irreversible fouling was achieved using NaOH + NaOCl solutions, with a subsequent citric acid cleaning step having a negative effect upon membrane pure water flux. The loss of pure water flux caused by citric acid cleaning was represented by an average $R_n/R_{n'}$ ratio of 1.51 over 10 cycles. The hydraulic resistance reduction recorded during cleaning with NaOH + NaOCl solutionsfollowed a first order removal model with time. A high coefficient of determination was found in every case (> 0.94).

The performance of ceramic membranes over multiple cycles is critical to industrial operation. The separation results are encouraging, showing good spore reduction with high solids transmission possible. Although cleaning regimes have still to be fully optimised, and are likely to be specific to individual processing environments, the importance of NaOH + NaOCl cleaning agents and the negative effect of citric acid have been highlighted. This

information provides a sound scientific basis for future trials, and industrial gum microfiltration operations.

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Figure 1

Typical resistance curve for full fouling and cleaning cycle. R_m, $R_{n'}$, R_n measured with pure water at 20° C, CFV 8.5 ms^{-1}, TMP 2.5 bar. R_t measured with 15wt% gum arabic solution at 50° C, CFV 8.5 ms^{-1}, TMP 2.9 bar. R_{fc}' and R_{fc} measured with NaOH+NaOCl and Citric acid solutions respectively at 60° C, CFV 11 ms^{-1}, TMP 2.5 bar.

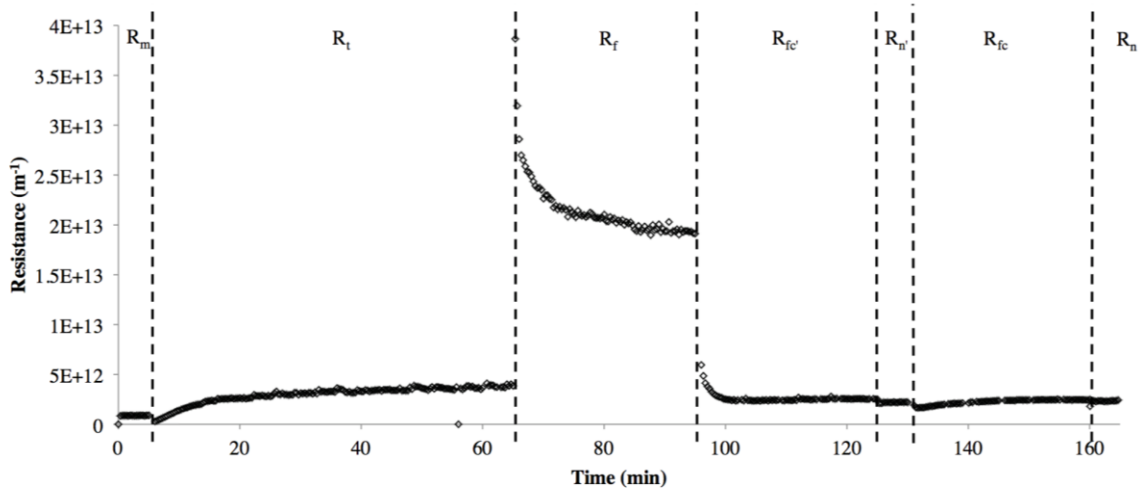

Figure 2

Variation of total resistance afer fouling (R_t) and total resistance after rinsing (R_f)

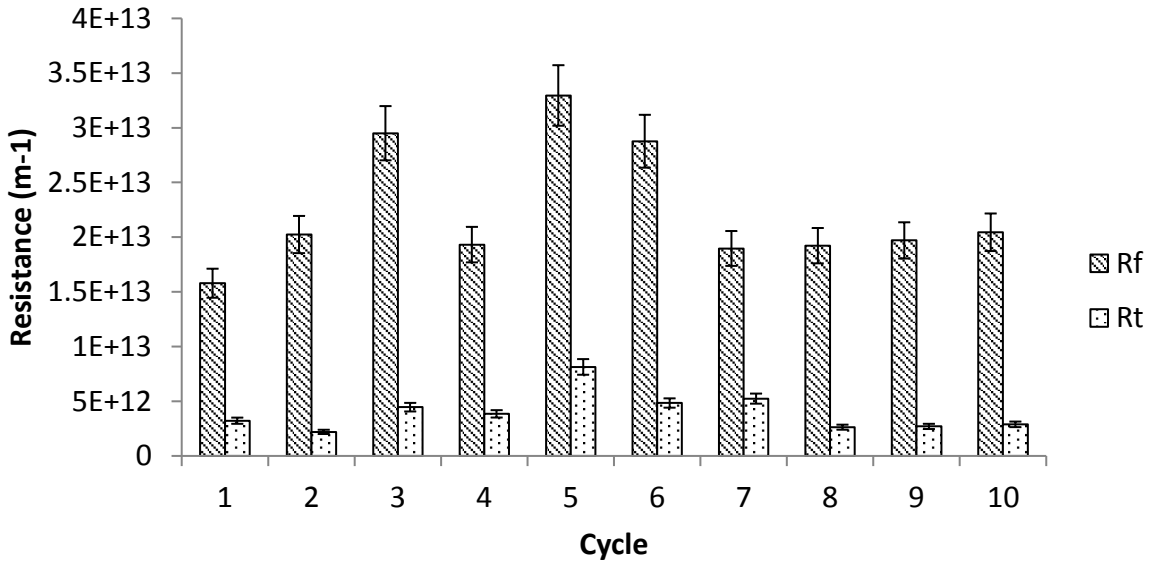

Figure 3

Variation in relative flux decline (RFD)

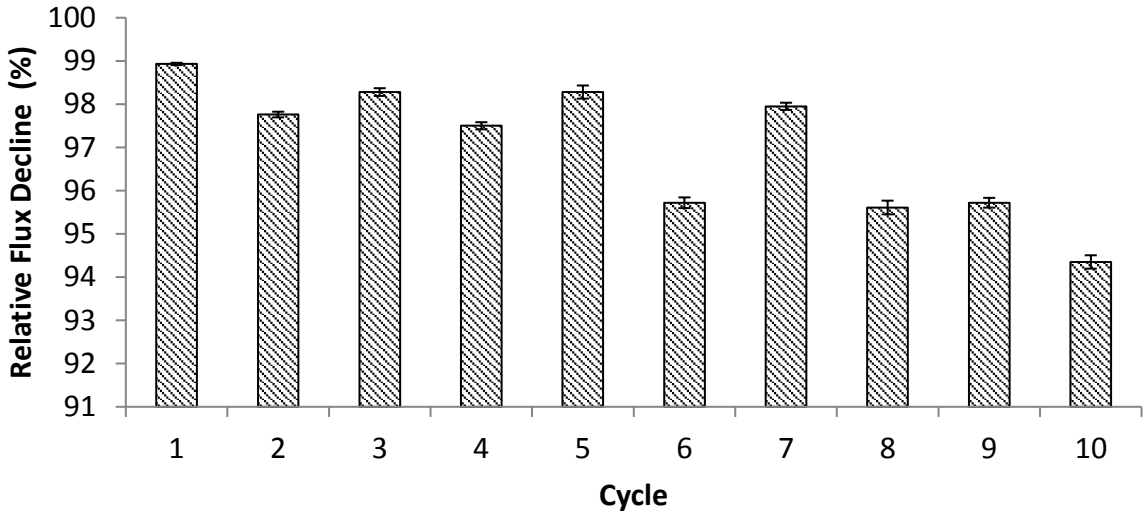

Figure 4

Solids retention (SR) against fouling resistance

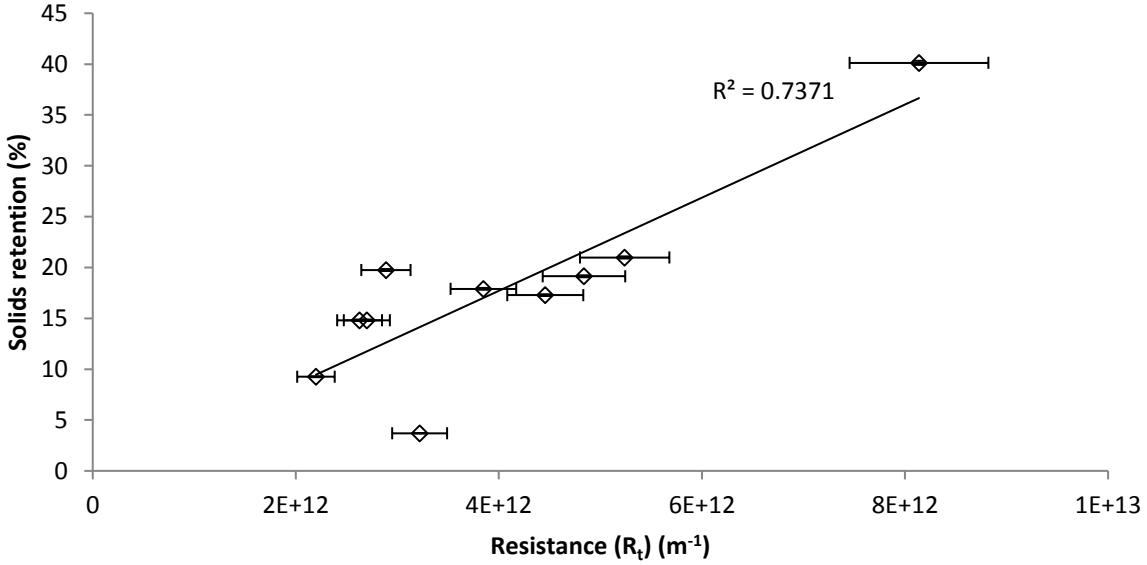

Figure 5

Variation in Flux Recovery (FR) over 10 cycles

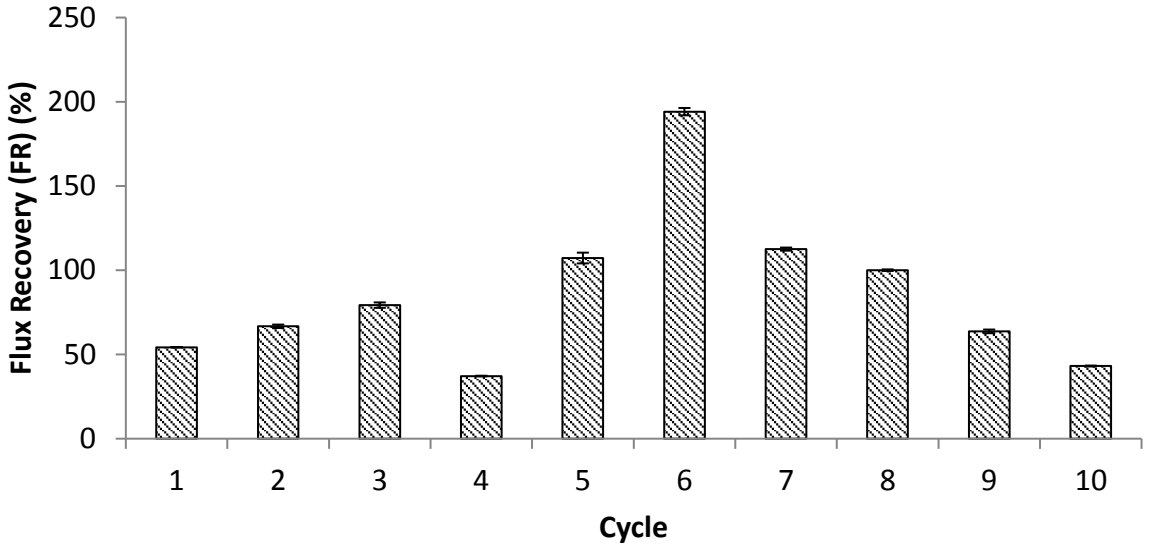

Figure 6

Hydraulic cleanliness criterion expressed after NaOH+Hypochlorite cleaning stage and after Citric acid cleaning stage

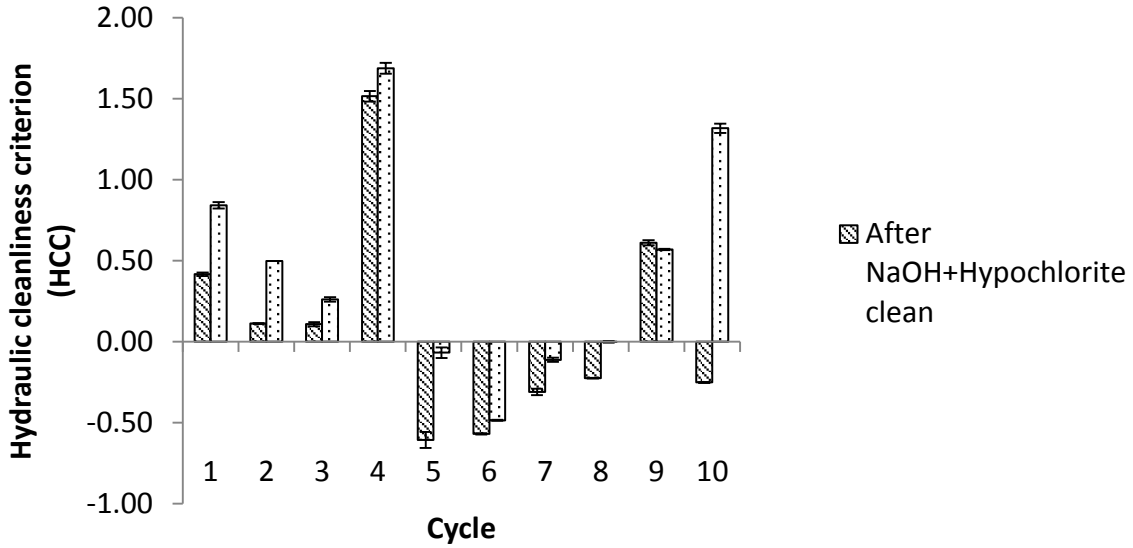

Figure 7

Ratio of resistance after Citric acid stage (R_n) to resistance after NaOH + Hypochlorite stage ($R_{n'}$)

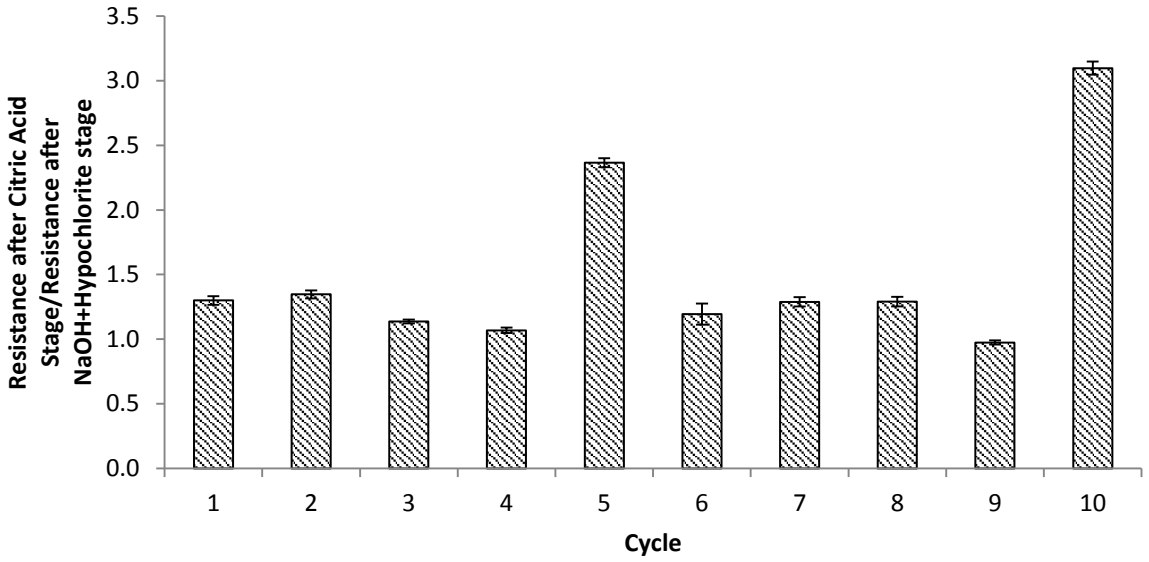

Figure 8

Example experimental data showing typical resistance decline during cleaning (R_{fc}). Zero order (n = 0) and first order (n = 1) model fits are shown overlaying the experimental data.